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Title: Project Overview: LA13-FY13-123-PD08

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PROJECT Overview

WebPMIS Number	LA13-FY13-123-PD08
Title	Improved Sample Utilization in TIMS Isotopic Ratio measurements via Refined Development and Application of Porous Ion Emitters
Years	FY13-FY14
Phenomenology	Mass Spectrometry
Fuel Cycle Stage	Other
Analytics techniques	Thermal Ionization Mass Spectrometry, Isotope Composition/Chronometry
Organizations (labs)	Los Alamos National Laboratory, Idaho National Laboratory
Key Staff	LANL: Floyd Stanley, Khal Spencer; INL: Matt Watrous, James Delmore
Summary	This project was a two year, LANL-led effort focused on extending the application of ion emitter sources in thermal ionization mass spectrometry based analysis of trace actinide systems. Particular emphasis was geared towards thorium isotope quantitation, both as a TIMS “worst case” scenario in terms of ionization efficiency and a desirable target for future chronometric measurements. Additional points of focus included source performance characteristics and identification of next-generation emitter strategies addressing remaining development challenges.
Classification Guides	
Author & email	Floyd Stanley, Ph.D. floyd@lanl.gov
Date/version	27-Sept-2017
References	• F.E. Stanley, K.J. Spencer, D.S. Schwartz, M.G. Watrous, J.E. Delmore. *J. Rad. Nuc. Chem.* 299 (2014) 1447-1452. • M.L. Baruzzini, H.L. Hall, K.J. Spencer, F.E. Stanley. *Int. J. Mass Spectrom.* (2017) Submitted. • J.M. Mannion, C.R. Shick Jr., G.A. Fugate, B.A. Powell, S.M. Husson. Anal. Chem. 89 (2017) 8638-8642.

PROJECT DETAILS

Goal: The goal of this research effort was to: 1) provide enhanced sample ionization strategies for thermal ionization mass spectrometry (TIMS) based measurement of trace actinide isotope composition, with an emphasis on thorium quantitation, 2) detail drivers of ionization and fractionation characteristics associated with porous ion emitters, and 3) conceptually identify next-generation strategies for improved emitter construction and performance in conjunction with the use of commercial instrumentation.

Motivation: TIMS methodology is widely used in the analysis of actinide systems relevant to various nuclear material arenas. However, the technique remains hindered by poor sample utilization, in the form of low ionization within the instrument source. Commonly employed, direct filament loading techniques, for example, are frequently associated with actinide ionization efficiencies of < 0.1% and the vast majority of sample remains unmeasured. This deficiency directly impacts the user community’s ability to measure trace level actinides in scenarios ranging from environmental measurement to chronometric evaluation of recently produced nuclear materials.

Technical Approach/Innovative Idea: The proposed effort focused on enhancing actinide sample utilization within commercially available TIMS instrumentation through the use of straightforward ion emitter construction and deposition procedures atop traditional filament assemblies. Execution in this manner eliminates traditional concerns of instrument modification/dedication which limit utility and throughput. Primary experimentation focused on thorium quantitation as a worst case scenario for thermal ionization and a straightforward means of amplify observed phenomena.

Project Objectives: The central objective of the work was geared towards extending porous ion emitter technologies/capabilities in TIMS to provide enhanced measurement of isotope composition in trace actinide systems.

Project sub-objectives included:

1. Initial interrogation of porous ion emitters as thermal ionization sources for the measurement of uranium progeny species; emphasis given to sample utilization efficiency, isotopic fractionation characteristics, and identification of performance drivers of value in future efforts.
2. Capitalizing on beneficial source characteristics to address current challenges in uranium chronometry, including initial application to historically difficult measurement systems.
3. Conceptual identification of next-generation schemes for constructing TIMS ion emitter sources that enhance utilization of a range of actinides, thereby reducing sample quantities needed to successfully obtain isotopic information, while maintaining applicability to “off-the-shelf” instrumentation.

Impact and remaining challenges: This effort provided proof of concept for the use of PIE sources in improved quantitation of thorium isotopic systems and the execution of chronometric characterization. Additionally, collected data provided insight into source mechanisms of functioning, potential utilization in addition actinide systems, and needed refinements to drive future improvements. The principal remaining challenge for this concept is the need to refine source construction methodologies to facilitate uniformity and ease in benchtop processing; these procedures will require further instrument driven interrogation to drive community application in actinide measurements.

Outcomes: The supported effort demonstrated PIE applicability in enhancing TIMS-based measurement of isotope ratios for a range of actinide systems (Th, Pu, Am). Furthermore, the executed work provided initial identification of drivers of ionization enhancement and proposed a next-generation ion emitter strategy (i.e. metal foam ion emitters) addressing concerns of heterogeneity within the constructed source.

No direct, NA-22 sponsored follow-on projects were identified during this review. However, several notable outcomes included: 1) Peer-reviewed publications, 2) Citation by others in the community pursuing related efforts under NA-22, and 3) Public presentation of findings. Items 1 and 2 above are captured in the supplied references section.